

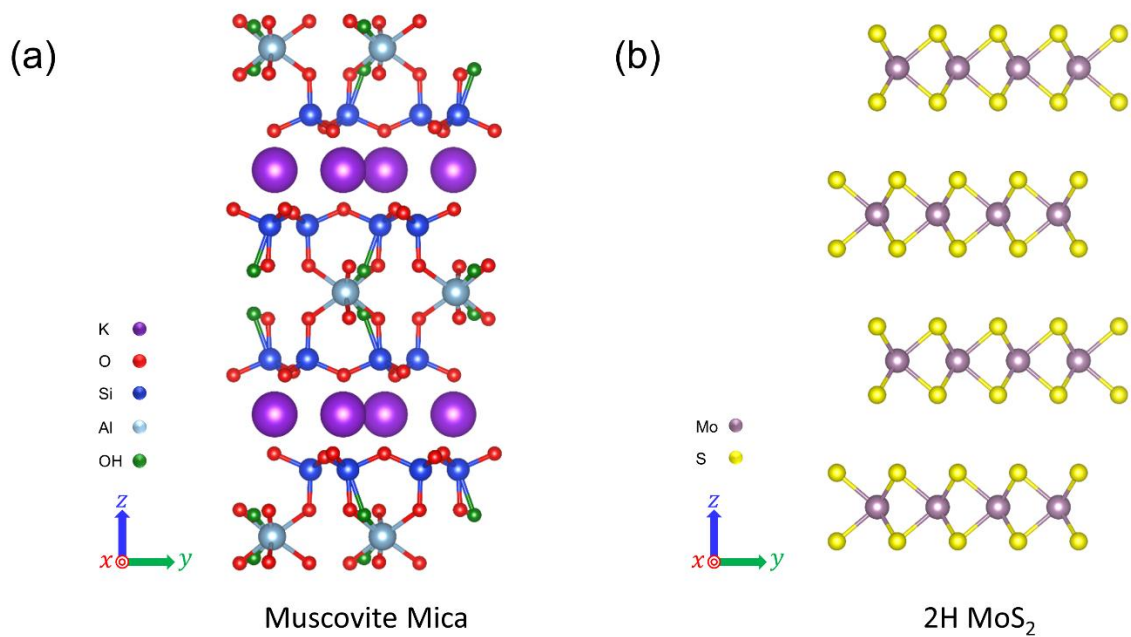
## Electronic Supplementary Information for

Unraveling the Influence of Substrate Surface and Temperature on Microstructural  
Evolution of Crystalline MoS<sub>2</sub> in Atomic Layer Deposition

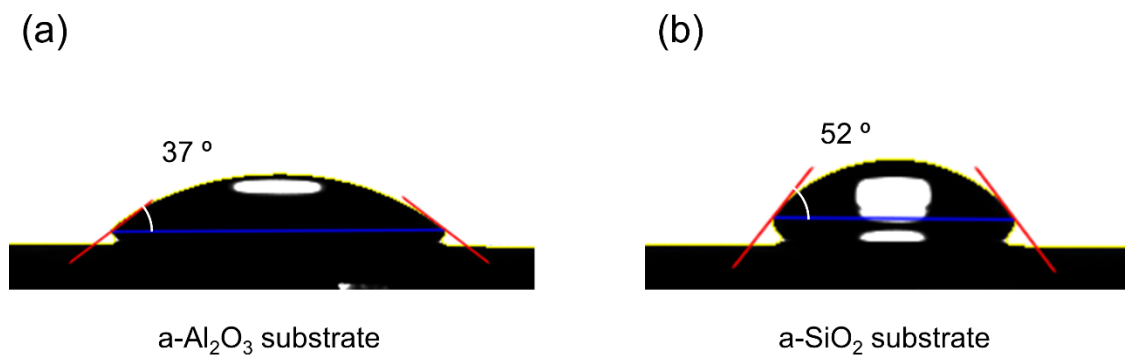
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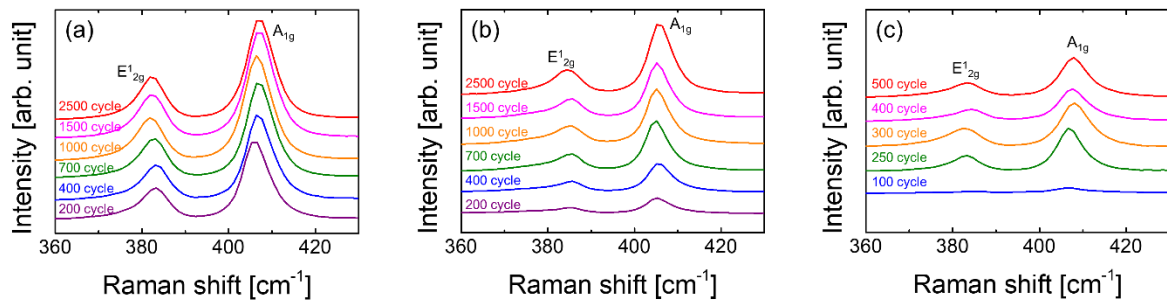
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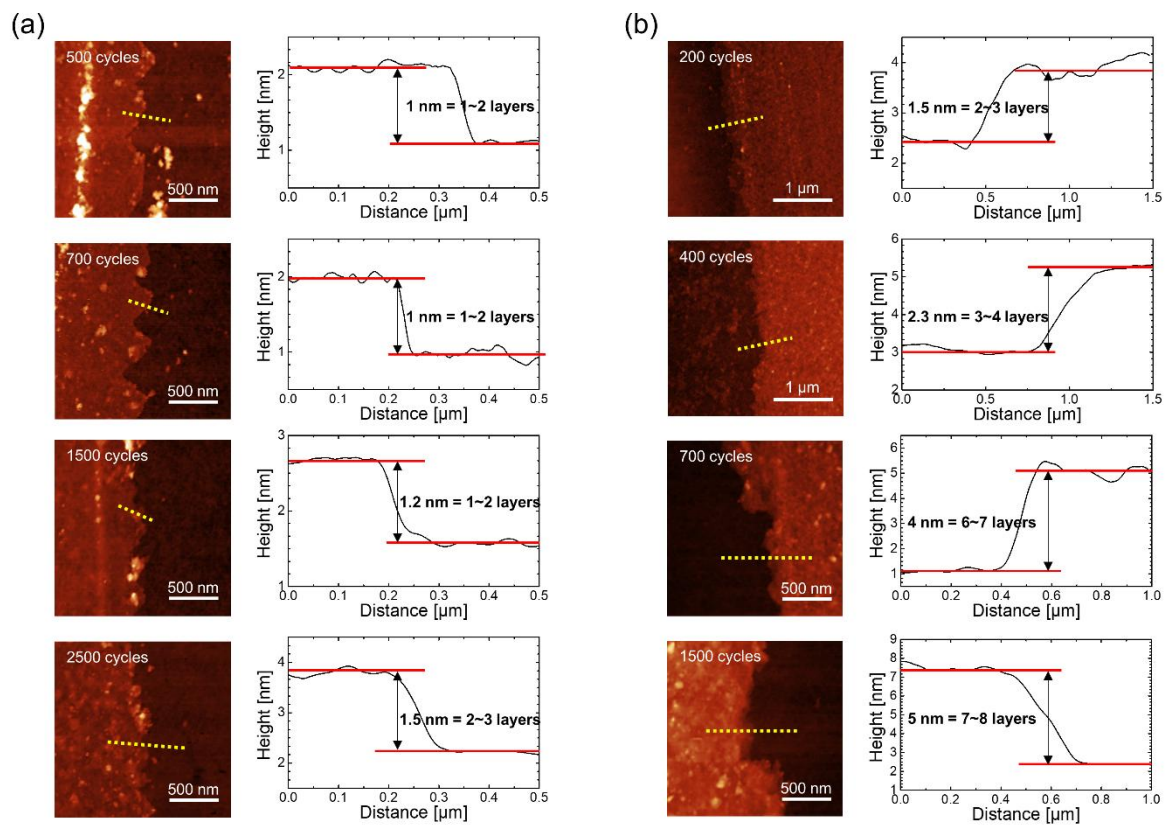
**Figure S1.** Schematics of (a) mica and (b) MoS<sub>2</sub> structures.



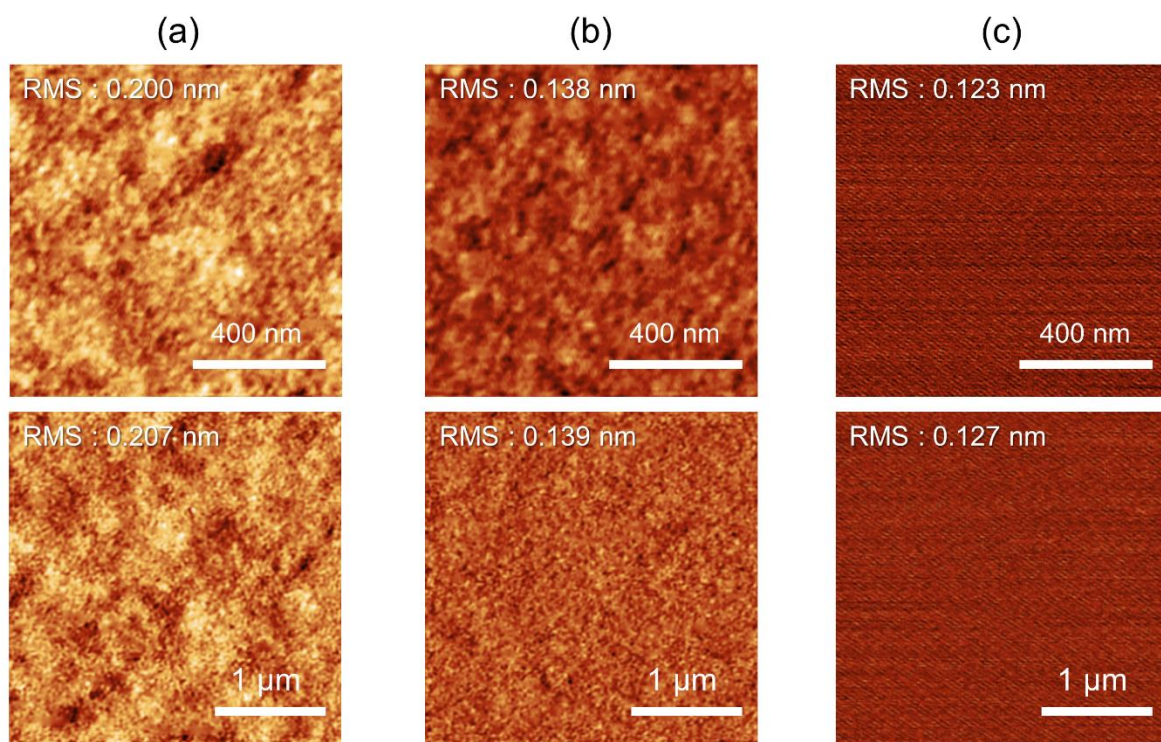
**Figure S2.** Contact angle measurements of (a) the ALD-grown Al<sub>2</sub>O<sub>3</sub> and (b) SiO<sub>2</sub> substrates



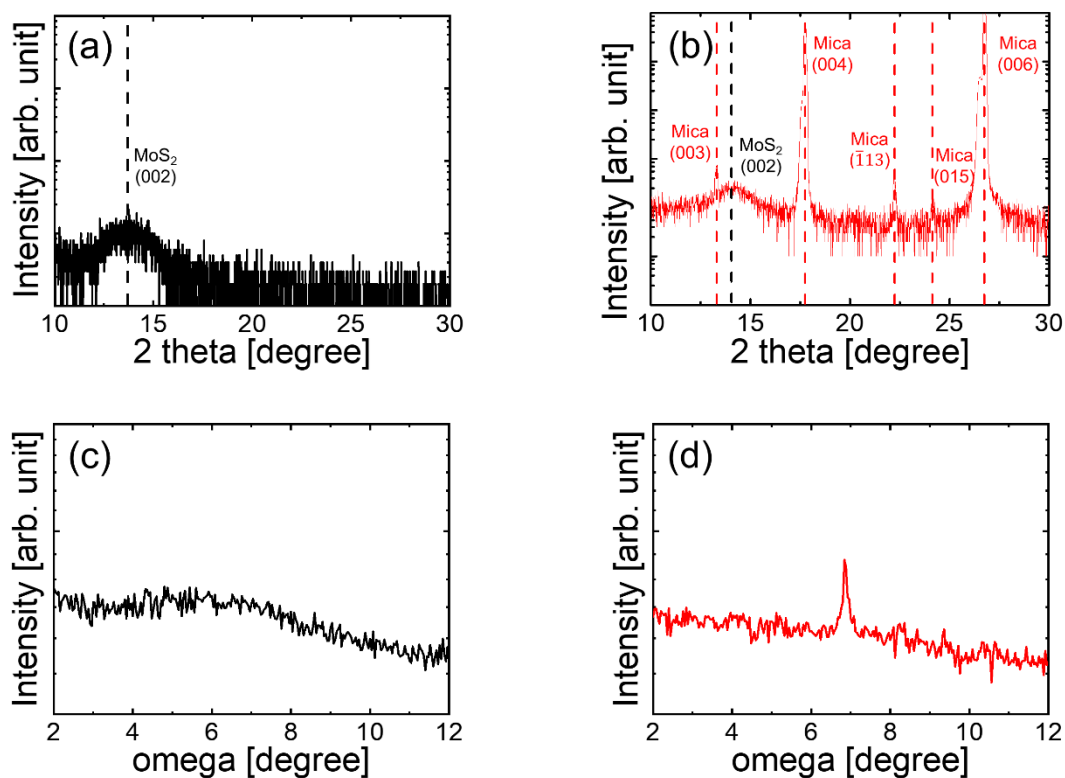
**Figure S3.** Raman spectra of MoS<sub>2</sub> grown on (a) Al<sub>2</sub>O<sub>3</sub>, (b) SiO<sub>2</sub>, and (c) mica at 650 °C



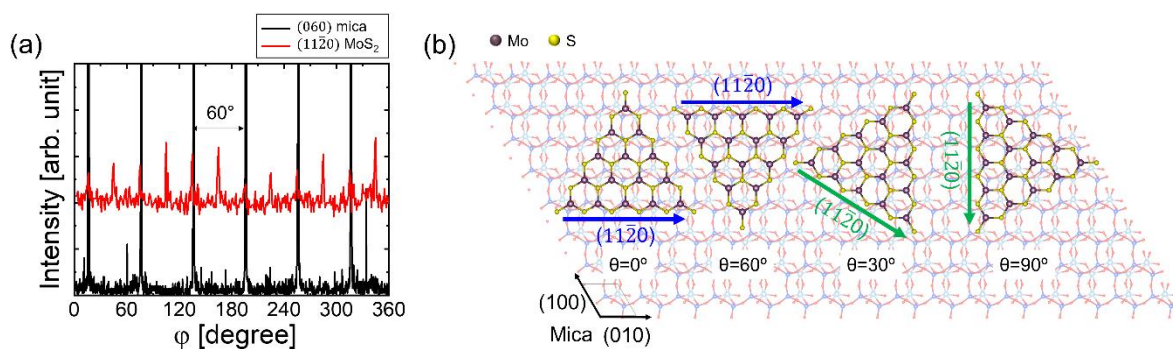
**Figure S4.** AFM images and height profiles of the MoS<sub>2</sub> grown on (a) SiO<sub>2</sub> and (b) Al<sub>2</sub>O<sub>3</sub> at 650 °C



**Figure S5.** AFM images of (a) SiO<sub>2</sub>, (b) Al<sub>2</sub>O<sub>3</sub>, and (c) mica substrates.



**Figure S6.**  $\theta$ - $2\theta$  theta XRD patterns of the MoS<sub>2</sub> grown on (a) the ALD-grown Al<sub>2</sub>O<sub>3</sub> and (b) the mica. Rocking curves of the MoS<sub>2</sub> grown on (c) the ALD-grown Al<sub>2</sub>O<sub>3</sub> and (d) the mica.



**Figure S7.** In-plane  $\phi$ -scans of the (060) mica and (11 $\bar{2}$ 0) MoS<sub>2</sub> reflections. (b) Schematic of possible coincidence site lattices for MoS<sub>2</sub> domains on mica.